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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF : EXAMINER: NAKARANI, DHIRAJLAL S.
CHIHARU OKAWARA, ET AL. :
SERIAL NO: 10/595,789 : ATTN: DECISIONS & CERTIFICATES OF
CORRECTION BRANCH
FILED: MAY 11, 2006 :
FOR: GAS BARRIER FILM : GROUP ART UNIT: 1794

RENEWED REQUEST FOR CERTIFICATE OF CORRECTION

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

The following is a Renewed Request for Certificate of Correction in U.S. patent application 10/595,786, now U.S. Patent 7,615,287.

Pursuant to 35 U.S.C. § 255 and 37 C.F.R. § 1.323, Applicants respectfully request that the U.S. Patent and Trademark Office issue a Certificate of Correction in the above-identified patent to correct obvious clerical errors that were inadvertently introduced in the previously submitted English language translation of the above-identified application.

Specifically, column 8, lines 31-32 and column 10, lines 1-2 of the specification, and column 17, lines 31-32 of claim 13, should be corrected to recite, in part, “inductively coupled plasma atomic emission spectroscopy (ICP-AES)” as set forth in the enclosed Form PTO-1050.

Support for these corrections is found at column 10, lines 1-4 of U.S. Patent 7,615,287, which recites that:

The analysis was performed through ***inductively coupled plasma Auger electron spectroscopy (ICP-AES)*** by means of a spectrometer (model ***ICAP-55***, product of ***NIPPON JARRELL-ASH***) (emphasis added).

The originally filed specification is alleged as failing to provide adequate written description for the recitation that “ICP-AES” refers to “inductively coupled plasma ***atomic emission*** spectroscopy” in column 17, lines 31-32 of claim 13, as well as column 8, lines 31-32 and column 10, lines 1-2 of the specification (emphasis added).

Pursuant to *In re Wertheim*, 541 F.2d 257, 265 (1976), the exact terms recited in the claimed invention need not be used *in ipsius verbis* or *in haec verba* in order to satisfy the written description requirement of 35 U.S.C. § 112, first paragraph. See also MPEP §§ 1302.01 and 2163.05(III). What is required is that the claimed invention must have been described with sufficient particularity such that a skilled artisan would recognize that the Applicants had possession of the claimed invention when the application was filed. See 35 U.S.C. § 112, first paragraph, and MPEP § 706.03(c).

With respect to the requested claim and specification amendments, the originally filed specification clearly discloses that the analysis was performed through ICP-AES spectroscopy by means of a spectrometer (model ICAP-55, product of NIPPON JARRELL-ASH). As shown in the enclosed journal article (Watanabe, M. and Narukawa, A., Determination of impurity elements in high purity graphite by inductively coupled plasma atomic emission spectrometry after microwave decomposition, *Analyst*, Vol. 125, pp. 1189-1191 (2000)), analysis by “inductively coupled plasma ***atomic emission*** spectrometry (ICP-AES)” is conducted using a “Nippon Jarrell-Ash Model ICAP-55 ICP-AES” spectrometer (See e.g., page 1189, column 1, lines 7-9, and page 1189, column 2, lines 3-4) (emphasis added).

Applicants respectfully submit that adequate support for the requested claim and specification amendments to correct obvious clerical errors that were inadvertently introduced in

the previously submitted English language translation of the above-identified application has clearly been provided by the express, implicit and inherent disclosure set forth in the originally filed specification, as evidenced hereinabove.

Since the specification describes the claimed invention in sufficient detail such that a skilled artisan would reasonably conclude that the inventors had possession of the claimed invention at the time of filing, the requested claim and specification amendments have not resulted in the introduction of new matter.

It is also believed that this claim amendment merely rectifies a formal matter with respect to an inadvertently introduced typographical error and therefore neither changes the scope of the claim nor requires reexamination of the claimed invention. Accordingly, issuance of the Certificate of Correction is respectfully requested.

In accordance with 37 C.F.R. § 1. 323, enclosed is the fee set forth in 37 C.F.R. § 1.20(a).

Respectfully submitted,

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UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 7,615,287
DATED: November 10, 2009
INVENTOR(S): Chiharu OKAWARA, et al.

It is certified that an error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

Column 8, lines 31-32, should read:

-- inductively coupled plasma atomic emission spectroscopy (ICP-AES) --

Column 10, lines 1-2, should read:

-- inductively coupled plasma atomic emission spectroscopy (ICP-AES) --

Column 17, Claim 13, lines 31-32, should read:

-- inductively coupled plasma atomic emission spectroscopy (ICP-AES) --

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Patent No. 7,615,287

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Determination of impurity elements in high purity graphite by inductively coupled plasma atomic emission spectrometry after microwave decomposition

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A high purity graphite powder was completely decomposed with a mixture of nitric and sulfuric acids using an open microwave digestion system. The excess nitric acid remaining in the solution was evaporated off using the same system. The sample solution was diluted with water and analyzed for the determination of impurity elements (Li, Na, K, Mg, Ca, Sr, Al, Fe, Ni, Cr, Ti, V) by inductively coupled plasma atomic emission spectrometry (ICP-AES). The analytical results for the elements in standard samples agreed well with those by other methods.

Graphite is used as a neutron reflector and/or moderator in nuclear reactors, a crucible for analytical instruments, an additive in various ceramics and also in other widespread fields. Thus, the establishment of precise methods for the determination of its impurities is very important, because the presence of impurities may affect the characteristics of the graphite even at trace levels. Prior to the measurement of the trace impurities by inductively coupled plasma atomic emission spectrometry (ICP-AES), the decomposition of the graphite is required without the introduction of any contamination.

Using an open wet digestion system, Hashitani *et al.*¹ decomposed graphite powder with a mixture of perchloric and periodic acids, however, their use of glass vessels for digestion severely contaminated the samples with both sodium and potassium. Watanabe and Inagawa² decomposed high purity graphite powder with perchloric, sulfuric and nitric acids in a quartz glass Erlenmeyer flask fitted with an air condenser. Watanabe and Takashima³ reported the decomposition in a sealed pressure vessel with a mixture of hydrochloric, nitric acids and water (1 + 1 + 1). Koshino and Narukawa⁴ decomposed graphite powder with a mixture of nitric and sulfuric acids at 250 °C for 72 h in a similar vessel. In these methods, the long time required for the decomposition was a problem. Furthermore, as a safety recommendation, the use of perchloric acid should be avoided.

This paper describes the complete decomposition of graphite with nitric and sulfuric acids using an open microwave digestion system under relatively moderate conditions with negligible contamination, followed by the determination of any metal impurities by ICP-AES.

for comparison under the experimental conditions reported previously.⁴

A Nippon Jarrell-Ash Model ICAP-55 ICP-AES (Nippon Jarrell-Ash, Kyoto, Japan) equipped with a Cetac (Omaha, NE, USA) U-5000AT ultrasonic nebulizer was used for the measurements of the impurities in the sample solutions under the operating conditions listed in Table 1.

Eppendorf (Hamburg, Germany) micropipettes (10–100 µl) were used for the handling of solutions. Nalgene (Nalge Company, Rochester, NY, USA) PMP measuring flasks (50 ml) and FEP beakers were used for the preparation of sample solutions. These vessels were cleaned by washing twice with a conc. hydrofluoric and nitric acid mixture, followed by rinsing with water. Millipore (Bedford, MA, USA) membrane filters, model no. GSWP 04700 (0.22 µm), were used to measure the residue after incomplete decomposition.

Reagents and samples

Spex (Metuchen, NJ, USA) Custom Standard Mix Solution XSTC-22 containing 1000 µg ml⁻¹ of Li, Na, K, Mg, Ca, Al, Fe, Ni, Cr, Ti and V and Spex Standard Solution of Sr (1000 µg ml⁻¹) were mixed and diluted with water to prepare the intermediate standard containing 100 µg ml⁻¹ of each element. Stepwise fractions (0–1000 µl) of this intermediate solution were diluted with 1.8 M sulfuric acid to prepare the working standards for constructing the calibration graphs for each element (0–2 µg ml⁻¹).

Table 1 The optimum operating condition for ICP-AES

Element	Wavelength /nm	Order	Sample flow rate/ml min ⁻¹	Rf power /kW
Li	670.784	1	2.3	0.8
Na	589.592	1	2.3	0.8
K	766.491	1	2.3	0.8
Mg	279.553	2	1.3	1.2
Ca	393.366	1	1.3	1.2
Sr	407.771	1	1.3	1.2
Al	396.152	1	1.3	1.2
Fe	238.204	3	1.3	1.2
Ni	221.647	3	1.3	1.2
Cr	205.552	3	1.3	1.2
Ti	334.941	1	1.3	1.2
V	309.311	1	1.3	1.2

Experimental

Apparatus and equipment

A Prolab (Paris, France) Model A301 microwave digestion system (micro-digester, maximum 200 W) was used for the decomposition of the graphite powder. A quartz-glass vessel (cat. no. 06 334 071, 100 ml) and quartz-glass air condenser (cat. no. 06 334 082) were attached to the system. The whole system was placed in a plastic box, which was ventilated with filtered air using a suction pump to prevent any corrosion from the exhaust gas or contamination from the environment. Decomposition in a sealed pressure vessel was also carried out

Nitric acid from Tama Chemical Tamapure AA-100 (Tokyo, Japan) and sulfuric acid from Kanto Cica-MERCK ultrapure (Tokyo, Japan) were used without further purification. Water from a Millipore Milli-Q SP TOC system was used throughout.

Three standard graphite powders (common graphite sample G3, CRM JAERI G5 and G6) were provided by Japan Atomic Energy Research Institute (JAERI).

Recommended procedures

A graphite powder of 0.25 g was placed into the 100 ml quartz-glass vessel with 5 ml of sulfuric and 5 ml of nitric acids. Attaching the air condenser, the vessel was placed in the micro-digester, and heated for 20 min at 60% microwave power, followed by a period of standing (for 1 min) and the addition of 4 ml of nitric acid. The sample was re-heated for 10 min at 65% power, then left for 1 min, after which 4 ml of nitric acid were added. Five more cycles were carried out with 10 min heating at 65% power, 1 min standing and the addition of 4 ml of nitric acid. Finally, the excess nitric acid was evaporated off by 15 min heating at the same power. The resultant clear solution was transferred into the PMP measuring flask (50 ml) and diluted to volume with water. The impurity elements were measured by ICP-AES.

Results and discussion

Decomposition of graphite

Using a sealed vessel, graphite (0.5 g) was completely decomposed with 20 ml of nitric and 4 ml of sulfuric acid.⁴ However, 0.25 g of graphite could not be decomposed with the same acids when using the proposed open digestion system, due to the evaporation of the nitric acid, which was then not available for digestion. The effects of the acid, the moderate heating and the repeated additions of nitric acid were considered for the proposed method. When heating for 10 min at 65% power, 4 ml of nitric acid evaporated from the decomposition system, the amount of nitric acid per heating cycle was therefore decided. The effect of the numbers of heating cycles on the decomposition rate of graphite is shown in Fig. 1. In this case,

0.25 g of JAERI G3 graphite was heated with 5 ml of sulfuric and 4 ml of nitric acids at 65% power. After every heating cycle, the solution was poured into an FEP beaker containing 50 ml of water and filtered with the membrane filter. The residue on the filter was dried at 60 °C for 2 h in an oven and the decomposition rate was calculated by weighing the residue. As can be seen in Fig. 1, the decomposition rate gradually increased with the number of heating cycles. A small amount of residue could be observed at the end of 4th cycle. The sample decomposition was completed by the 5th cycle.

By varying the amounts of graphite from 0.1 to 0.5 g, the decomposition rate was investigated with the recommended procedure. As shown in Fig. 2, up to 0.3 g of graphite was completely decomposed.

As graphite is converted into carbon dioxide and the concentrations of the impurity elements in the measured solutions are in the $\mu\text{g ml}^{-1}$ range, the working standard solutions could contain all the target elements and 1.8 M sulfuric acid could be used for matrix matching between the sample and standard solutions.

The recoveries for the impurity elements in the present method were from 99% to 106%, confirming no volatilization-loss and/or contamination of the target elements. These values were obtained by adding 12.5 μg of the intermediate standards to a blank.

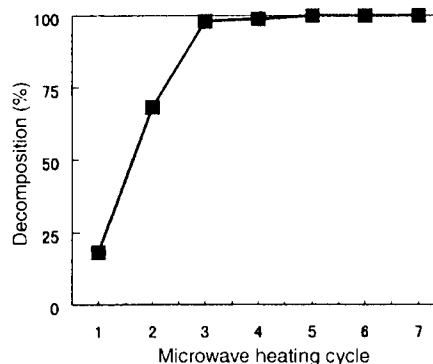


Fig. 1 Effect of repeated heating cycles on the decomposition of the graphite powder. Sample (G3 0.25 g) was decomposed on the basis of the recommended procedure.

Table 2 Analytical results of impurity elements in various graphite powders in ($\mu\text{g g}^{-1}$)

Sample and method	Li	Na	K	Mg	Ca	Sr	Al	Fe	Ni	Cr	Ti	V
G 3												
Collaborative study ^{5,6}				0.1 + 0.01	209 + 8	4.0 + 0.6	1.0 + 0.3	41.4 + 2.7	27.1 + 2.2	1.3 + 0.29	5.2 + 0.3	42.3 + 2.5
Acid decomposition												
Sealed vessel	< 0.1	0.6 + 0.1	0.3 + 0.1	0.1 + 0.1	219 + 4	4.5 + 0.1	0.5 + 0.1	46.6 + 0.7	31.5 + 0.3	1.2 + 0.1	5.7 + 0.1	47.5 + 0.1
Open vessel ¹²					222 + 5	4.8 + 0.2		46.4 + 1.1	30.0 + 1.5		5.2 + 0.6	47.1 + 1.0
Present method	< 0.2	0.4	0.6	0.2	220	4.5	0.7	45.4	30.7	1.2	5.6	46.7
	< 0.2	0.8	0.7	0.1	222	4.7	1.0	45.5	31.1	1.2	5.8	47.5
G 5												
Collaborative study				< 0.1	< 0.2	< 0.1	< 0.2	< 0.5	< 0.1	< 0.1	< 0.1	< 0.1
Acid decomposition												
Sealed vessel	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Present method	< 0.2	< 0.2	< 0.5	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
	< 0.2	< 0.2	< 0.5	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
G 6												
Collaborative study				0.2 ^a	36.2 + 1.7	0.57 + 0.05	2 ^a	10.9 + 1.5	0.27 + 0.07	0.3 ^a	18.4 + 0.8	2.9 + 0.2
Acid decomposition												
Sealed vessel	0.2 + 0.1	0.7 + 0.1	1.2 + 0.1	0.2 + 0.1	38.7 + 0.7	0.6 + 0.1	1.8 + 0.1	11.5 + 0.1	0.3 + 0.1	0.4 + 0.1	19.7 + 0.4	3.1 + 0.1
Open vessel					38.4 + 1.4	0.87 + 0.18		10.5 + 0.9			19.8 + 0.6	3.3 + 0.1
Present method	< 0.2	0.9	1.4	0.2	39.3	0.6	1.5	10.4	0.3	0.5	19.3	3.1

^a Ref. 6, reference value.

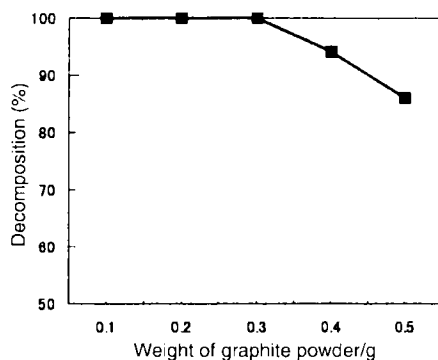


Fig. 2 Effect of the sample weight on the decomposition rate of the graphite powder.

Analytical results

The analytical results for the impurity elements in three common graphite samples of G3, and CRM JAERI G5 and G6 are summarized in Table 2, together with those by other

methods. The analytical values obtained for the proposed method agree quite well with those by the others. As the G5 sample is extremely pure, the contents of the impurity elements are less than the detection limits of the proposed method. The detection limits are defined as three times the standard deviation of five procedure blanks.

References

- 1 H. Hashitani, H. Yoshida, T. Adachi and K. Izawa, *Bunseki Kagaku*, 1986, **35**, 911.
- 2 K. Watanabe and J. Inagawa, *Analyst*, 1996, **121**, 623.
- 3 K. Watanabe and K. Takashima, *Abstracts of the 51st Symposium on Analytical Chemistry*, The Japan Society for Analytical Chemistry, Tokyo, 1990, p. 523.
- 4 Y. Koshino and A. Narukawa, *Analyst*, 1993, **118**, 827.
- 5 K. Watanabe, K. Takashima, S. Tamura and Y. Toita, *Abstracts of the 51st Symposium on Analytical Chemistry*, The Japan Society for Analytical Chemistry, Tokyo, 1990, p. 431.
- 6 *Chemical Analysis of High Purity Graphite: Preparation of Certified Reference Materials and Development of Analytical Methods*, Japan Atomic Energy Research Institute, Japan, 1993 Report JAERI-M 93-013.

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2 文字コード及び他の略語については、定期発行される各 PCT ガゼットの巻頭に掲載されている「コードと略語のガイダンスノート」を参照。

(54) Title: GAS BARRIER FILM

(54) 発明の名称: ガスバリア性フィルム

(57) Abstract: Disclosed is a film having high gas barrier properties immediately after it is produced. The film has higher gas barrier properties than the conventional films, and can be produced at low cost by a process having the minimum steps. The gas barrier film is obtained by forming an inorganic thin film on at least one side of a thermoplastic polymer film and applying a solution over the organic thin film side, which solution contains at least one kind of low molecular weight electrolyte ions having a molecular weight of not more than 1000 and selected from the group consisting of alkali metal ions, alkaline earth metal ions and ammonium ions. The total concentration of ions in the solution is not less than 1×10^{-5} mol/L and less than 10 mol/L, and also less than the saturated solution concentration.

(57) 要約: 製造直後から高いガスバリア性を示し、更に従来に比べ高いガスバリア性を示すフィルムを、最小限の工程及び低コストで提供する。熱可塑性高分子フィルムの少なくとも一方の面に無機薄膜を設け、該無機薄膜の側に、アルカリ金属イオン、アルカリ土類金属イオン、及びアンモニウムイオンからなる群より選ばれた分子量 1000 以下の低分子量電解質のイオンの少なくとも 1 種を含み、該イオンの総濃度が 1×10^{-5} mol/L 以上、乃至 10 mol/L 未満且つ飽和溶液濃度未満の溶液を塗布してなるガスバリア性フィルム。

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は生分解性樹脂が好ましい。

[0022] 本発明においては、無機薄膜を形成した後、或いはアルカリ金属イオン、アルカリ土類金属イオン、及びアンモニウムイオンからなる群より選ばれた分子量1000以下の低分子量電解質のイオンの少なくとも1種を含む溶液を塗布した後、或いは樹脂溶液及び／又は金属酸化物ゾル溶液を塗布した後に、ガスバリア性、膜質及び塗布層質の安定化等の点から加熱処理を施すことが好ましい。

加熱処理は、ガスバリア性フィルムを構成する要素の種類や厚さなどによりその条件が異なるが、必要な温度、時間を維持できる方法であれば特に限定されない。例えば、必要な温度に設定したオープンや恒温槽で保管する方法、熱風を吹き付ける方法、赤外線ヒーターで加熱する方法、ランプで光を照射する方法、熱ロールや熱版と接触させて直接的に熱を付与する方法、マイクロ波を照射する方法などが使用できる。また、取り扱いが容易な大きさにフィルムを切断してから加熱処理しても、フィルムロールのままに加熱処理してもよい。更に必要な時間と温度が得られる限りにおいては、コーター、スリッター等のフィルム製造装置の一部分に加熱装置を組み込み、製造過程で加熱を行うこともできる。

[0023] 加熱処理の温度は、使用する基材、プラスチックフィルム等の融点以下の温度であれば特に限定されないが、熱処理の効果が発現するために必要な処理時間を適度に設定できることから60℃以上であることが好ましく、更に70℃以上で行うことが好ましい。加熱処理温度の上限は、ガスバリア性フィルムを構成する要素の熱分解によるガスバリア性の低下を防止する観点から、通常200℃、好ましくは160℃である。処理時間は、加熱処理温度に依存し、処理温度が高い程、短くすることが好ましい。例えば、加熱処理温度が60℃の場合、処理時間は3日～6ヶ月程度、80℃の場合、処理時間は3時間～10日程度、120℃の場合、処理時間は1時間から1日程度、150℃の場合、処理時間は3～60分程度であるが、これらは単なる目安であって、ガスバリア性フィルムを構成する要素の種類や厚さ等により適宜調整することができる。

[0024] 本発明のガスバリア性フィルムにおいては、優れたガスバリア性効果を発現する点から、誘導結合プラズマ原子発光分析(ICP-AES)を用いて測定した、フィルム面積1.00m²における無機薄膜中、好ましくは酸化珪素を含む無機薄膜中のアルカリ

無機薄膜中の何れかの深さ領域において、上記C/B範囲値をとることが本発明のガスバリア性フィルムにおいて好ましい。イオン強度Bに ^{30}Si イオン強度を用いる理由は、酸化珪素を含む薄膜の構成元素Siのうち、 ^{30}Si は他の同位体 ^{28}Si 、 ^{29}Si に比べ検出強度が飽和する可能性がなく、ピーク干渉の影響の受け難いからである。

- [0027] 本発明においては、上記A/Bが100以下である場合は、析出による外観不良や薄膜にダメージが生じにくく、一方、0.20以上であれば、アルカリ金属イオン、アルカリ土類金属イオン、アンモニウムイオンの薄膜への吸着が十分になされ、優れたガスバリア性効果が得られる。また、C/Bが0.50以下であれば、薄膜中の酸化珪素においてSi-O鎖が十分に存在し、薄膜の機械強度に優れることから好ましく、一方、C/Bが0.04以上であれば、従来の無機薄膜ガスバリア性フィルムに比べ特性上ガス透過を阻害する点において優れる。

実施例

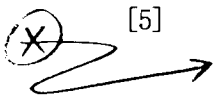
- [0028] 以下、本発明を実施例により具体的に説明するが、本発明は以下の例に限定されるものではない。なお、以下の実施例におけるフィルムの評価方法は、次の通りである。

< 薄膜中の金属元素分析(ICP) >

前処理は、面積 1.00m^2 分の薄膜フィルムを、長さ50cm幅5cm毎に切り、プラスチックボトル中でイオン交換水で洗浄する。そして、そのフィルムを別のプラスチックボトル中でフッ酸15mLと硝酸5mLとイオン交換水30mLの混合溶液に入れ薄膜を溶解し(溶解後の液を溶液(ア)とする)、次いでそのフィルムを別のプラスチックボトル中でイオン交換水で洗浄する(洗浄後の液を溶液(イ)とする)。溶液(ア)、(イ)を共に硫酸1mLを添加した白金坩堝中でドライアップし、その後塩酸2mLを添加、加温して白金坩堝中の析出した無機物を溶解する(溶解後の液を溶液(ウ)とする)。溶液(ウ)を25mLポリメスフラスコでイオン交換水で定容し、試料溶液とし分析した。フッ酸、硝酸、硫酸、塩酸は、電子工業用EL試薬または原子吸光用試薬を用いた。プラスチックボトルは、金属元素が溶出しないものを用いた。

⑧ { 分析は、誘導結合プラズマ原子発光分析(ICP-AES)で行い、装置はNIPPON J ARRELL-ASH社製「ICAP-55型」を用いた。測定値を表1-1及び表1-2に示す

請求の範囲

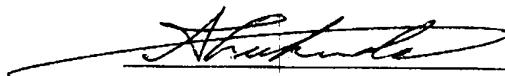
- [1] 熱可塑性高分子フィルム of の少なくとも一方の面に無機薄膜を設け、該無機薄膜の側に、アルカリ金属イオン、アルカリ土類金属イオン、及びアンモニウムイオンからなる群より選ばれた分子量1000以下の低分子量電解質のイオンの少なくとも1種を含み、該イオンの総濃度が $1 \times 10^{-5} \text{ mol/L}$ 以上、乃至 10 mol/L 未満且つ飽和溶液濃度未満の溶液を塗布してなるガスバリア性フィルム。
- [2] アルカリ金属イオン、アルカリ土類金属イオン、及びアンモニウムイオンからなる群より選ばれた分子量1000以下の低分子量電解質のイオンの少なくとも1種のイオンの総濃度が、 $1 \times 10^{-4} \text{ mol/L}$ 以上、乃至 $1 \times 10^{-1} \text{ mol/L}$ 未満且つ飽和溶液濃度未満である請求項1記載のガスバリア性フィルム。
- [3] 熱可塑性高分子フィルム of の少なくとも一方の面に無機薄膜を設け、該無機薄膜の側に、アルカリ金属イオン、アルカリ土類金属イオン、及びアンモニウムイオンからなる群より選ばれた分子量1000以下の低分子量電解質のイオンの少なくとも1種を含む溶液を、フィルム面積 1.00 m^2 あたりの上記イオンの総重量が $1.0 \mu\text{g}$ 以上、乃至 30 g 以下となるように塗布してなるガスバリア性フィルム。
- [4] 無機薄膜が酸化珪素を含む薄膜である請求項1〜3のいずれか1項に記載のガスバリア性フィルム。
-  [5] 熱可塑性高分子フィルム of の少なくとも一方の面に無機薄膜を有し、誘導結合プラズマ原子発光分析(ICP-AES)を用いて測定した、フィルム面積 1.00 m^2 における上記薄膜中のアルカリ金属及びアルカリ土類金属の総濃度が、 $2.0 \mu\text{g}$ 以上、乃至 $1000 \mu\text{g}$ 以下であるガスバリア性フィルム。
- [6] 飛行時間型二次イオン質量分析計(TOF-SIMS)による酸化珪素を含む薄膜中のアルカリ金属イオン、アルカリ土類金属イオン、及びアンモニウムイオンの総和イオン強度Aが、 ^{30}Si イオン強度Bに対し、 $0.20 \leq A/B \leq 100$ である請求項4記載のガスバリア性フィルム。
- [7] 飛行時間型二次イオン質量分析計(TOF-SIMS)による酸化珪素を含む薄膜中の $^{30}\text{SiOH}$ イオン強度Cが、 ^{30}Si イオン強度Bに対し、 $0.04 \leq C/B \leq 0.50$ である請求項4記載のガスバリア性フィルム。

CERTIFICATION

I, Akihito Fukuda, Bridgestone Toranomon Building 6F, No. 25-2, Toranomon 3-chome, Minato-ku, Tokyo, 105-0001, Japan do hereby certify that I am conversant with the English and Japanese languages and am a competent translator thereof. I further certify that to the best of my knowledge and belief the attached English translation is a true and correct translation of the paragraphs [0024] and [0028], and claim 5 of PCT application No. PCT/2004/017680 filed on November 29, 2004.

November 2, 2010

Date



Akihito Fukuda

Paragraph [0024]

In the gas barrier film of the present invention, in order to exhibit excellent gas barrier performance, the inorganic thin film (preferably the inorganic film containing silicon oxide) preferably has a total alkali metal ion and alkaline earth metal ion concentration in a film area of 1.00 m², as determined through inductively coupled plasma atomic emission spectroscopy (ICP-AES), of 2.0 µg to 1,000 µg, more preferably 2.0 µg to 500 µg, particularly preferably 5.0 µg to 200 µg.

The silicon oxide-containing inorganic thin film of the gas barrier film of the present invention preferably has a ratio of total ion intensity A to ion intensity B (A/B), as determined by means of a time-of-flight secondary ion mass spectrometer (TOF-SIMS), of $0.20 \leq A/B \leq 100$, more preferably $0.25 \leq A/B \leq 10$, wherein ion intensity A represents the sum of peak intensities attributed to total alkali metal ions, alkaline earth metal ions, and ammonium ions contained in the thin film, and ion intensity B represents a peak intensity attributed to ³⁰Si.

Furthermore, the silicon oxide-containing inorganic thin film of the gas barrier film of the present invention preferably has a ratio of ion intensity C to ion intensity B (³⁰Si) (C/B), as determined by means of a TOF-SIMS, of $0.040 \leq C/B \leq 0.50$, more preferably $0.050 \leq C/B \leq 0.10$, wherein ion intensity C represents a peak intensity attributed to ³⁰SiOH.

In the terms "ion intensity A" (the sum of peak intensities attributed to total alkali metal ions, alkaline earth metal ions, and ammonium ions) and "ion intensity B" (a peak intensity attributed to ³⁰Si), these ions contained in the inorganic thin film are detected as corresponding ion species during TOF-SIMS analysis.

Paragraph [0028]

The present invention will next be described in detail by way of examples, which should not be construed as limiting the invention thereto. In the Examples, film evaluation was performed as follows.

<Metal element analysis of thin film (ICP)>

As a preliminary treatment, a thin film (film area: 1.00 m²) was cut into pieces (length: 50 cm, width: 5 cm), and these pieces were washed with ion-exchange water in a plastic bottle. The film pieces were dissolved in a solution containing hydrofluoric acid (15 mL), nitric acid (5 mL), and ion-exchange water (30 mL) placed in another plastic bottle (the solution after dissolution is referred to as solution (A)). Subsequently, remaining film pieces were washed with ion-exchange water in another plastic bottle (the wash liquid is referred to as solution (B)). Solutions (A) and (B) were transferred to a platinum crucible containing sulfuric acid (1 mL) so as to dry the solutions. Hydrochloric acid (2 mL) was added to the dried product, followed by heating, whereby inorganic substances deposited in the platinum crucible were dissolved (the solution after dissolution is referred to as solution (C)). Solution (C) was diluted in a 25-mL plastic measuring flask with ion-exchange water to adjust the concentration to a predetermined value, and the solution served as an analyte solution. Hydrofluoric acid, nitric acid, sulfuric acid, and hydrochloric acid used here were all EL reagents for use in the electronics industry or reagents for atomic absorption spectrometry. The employed plastic bottles were made of a material which does not release metallic elements.

The analysis was performed through inductively coupled plasma atomic emission spectroscopy (ICP-AES) by means of a spectrometer (model ICAP-55, product of NIPPON JARRELL-ASH). The results of the analysis are shown in Tables 1-1 and 1-2.

Claim 5

A gas barrier film comprising a thermoplastic polymer film, and an inorganic thin film provided on at least one surface of the thermoplastic polymer film, wherein the thin film has a total alkali metal ion and alkaline earth metal ion concentration in a film area of 1.00 m², as determined through inductively coupled plasma atomic emission spectroscopy (ICP-AES), of 2.0 µg to 1,000 µg.